

CLAIMS

1. A positive electrode active material for a lithium secondary battery, which comprises a lithium-cobalt composite oxide represented by the formula $\text{Li}_p\text{Co}_x\text{M}_y\text{O}_z\text{F}_a$ (wherein M is a transition metal element other than Co or an alkaline earth metal element, $0.9 \leq p \leq 1.1$, $0.980 \leq x \leq 1.000$, $0 \leq y \leq 0.02$, $1.9 \leq z \leq 2.1$, $x+y=1$ and $0 \leq a \leq 0.02$) and comprising a mixture containing substantially spherical first particles of lithium-cobalt composite oxide having such a sharp particle size distribution that the volume basis cumulative size D10 is at least 50% of the average particle size D50, and the volume basis cumulative size D90 is at most 150% of the average particle size D50, and second particles of lithium-cobalt composite oxide filling the space among the above lithium-cobalt composite oxide particles, in a mass ratio of first particles/second particles of from 1/2 to 9/1.
2. The positive electrode active material according to Claim 1, wherein in the formula, M is at least one member selected from the group consisting of Ti, Zr, Hf, V, Nb, Ta, Mn, Mg, Ca, Sr, Ba and Al.
3. The positive electrode active material according to Claim 1 or 2, wherein the average particle size D50 is from 5 to 15 μm , the specific surface area is from 0.3 to 0.7 m^2/g , the half value width of the diffraction peak on (110) plane at $2\theta=66.5 \pm 1^\circ$ is from 0.07 to 0.14° as measured by X-ray diffraction using $\text{CuK}\alpha$ as a radiation

source, and the press density is from 3.1 to 3.4 g/cm³.

4. The positive electrode active material according to any one of Claims 1 to 3, wherein the first particles are large particles having an average particle size D50 of
5 from 7 to 20 μ m, and the second particles are small particles having an average particle size of from 10 to 30% of D50 of the first particles.

5. The positive electrode active material according to any one of Claims 1 to 4, wherein the first particles
10 have a press density of from 2.9 to 3.2 g/cm³, and the second particles have a press density of from 2.7 to 3.1 g/cm³.

6. A process for producing the positive electrode active material for a lithium secondary battery as
15 defined in any one of Claims 1 to 3, which comprises firing, as a cobalt source, a mixture of substantially spherical large particle size cobalt hydroxide or tricobalt tetraoxide having such a sharp particle size distribution that the average particle size D50 is from 7
20 to 20 μ m, the average particle size D10 is at least 50% of the average particle size D50 and the average particle size D90 is at most 150% of the average particle size D50, and small particle size cobalt hydroxide or tricobalt tetraoxide having an average particle size D50 of from 10
25 to 30% of the average particle size D50 of the large particles, in a proportion of from 9:1 to 1:2 as the cobalt atomic ratio, at a temperature of from 700°C to

1050°C in an oxygen-containing atmosphere.

7. The production process according to Claim 6, wherein the large particle size cobalt hydroxide or tricobalt tetraoxide has a press density of from 1.7 to 3.0 g/cm³,
5 and the small particle size cobalt hydroxide or tricobalt tetraoxide has a press density of from 1.7 to 3.0 g/cm³.

8. The production process according to Claim 6 or 7, wherein each of the large particle size cobalt hydroxide or tricobalt tetraoxide and the small particle size
10 cobalt hydroxide or tricobalt tetraoxide has a specific surface area of from 2 to 20 m²/g.

9. The production process according to any one of Claims 6 to 8, wherein the large particle size or small particle size cobalt hydroxide has a half value width of
15 the diffraction peak on (001) plane at $2\theta=19\pm1^\circ$ of from 0.18 to 0.35° and a half value width of the diffraction peak on (101) plane at $2\theta=38\pm1^\circ$ of from 0.15 to 0.35°, in an X-ray diffraction spectrum using CuK α -ray.

10. A process for producing the positive electrode
20 active material for a lithium secondary battery as defined in any one of Claims 1 to 3, which comprises firing, as a cobalt source, a mixture of substantially spherical cobalt hydroxide or tricobalt tetraoxide having such a sharp particle size distribution that the average
25 particle size D50 is from 7 to 20 μm , the average particle size D10 is at least 50% of the average particle size D50, the average particle size D90 is at most 150%

of the average particle size D50, and the average particle size of secondary particles formed by agglomeration of primary particles is from 8 to 20 μm , and cobalt oxyhydroxide having an average particle size of secondary particles formed by agglomeration of primary particles of from 7 to 20 μm , in a proportion of from 5:1 to 1:5 as the cobalt atomic ratio, at a temperature of from 700°C to 1050°C in an oxygen-containing atmosphere.

11. The production process according to Claim 10, wherein the cobalt oxyhydroxide has a half value width of the diffraction peak on (220) plane at $2\theta=31\pm1^\circ$ of at least 0.8° and a half value width of the diffraction peak on (311) plane at $2\theta=37\pm1^\circ$ of at least 0.8° , in an X-ray diffraction spectrum using $\text{CuK}\alpha$ -ray, and has a specific surface area of from 10 to 80 m^2/g .

12. The production process according to Claim 10 or 11, wherein as the cobalt hydroxide, substantially spherical cobalt hydroxide having a half value width of the diffraction peak on (001) plane at $2\theta=19\pm1^\circ$ of at least 0.15° and a half value width of the diffraction peak on (101) plane at $2\theta=38\pm1^\circ$ of at least 0.15° , in an X-ray diffraction spectrum using $\text{CuK}\alpha$ -ray, and having a specific surface area of from 2 to 30 m^2/g , is used.

13. The production process according to any one of Claims 10 to 12, wherein the tricobalt tetraoxide has a half value width of the diffraction peak on (220) plane at $2\theta=31\pm1^\circ$ of at least 0.08° and a half value width of

the diffraction peak on (311) plane at $2\theta=37\pm1^\circ$ of at least 0.10° , in an X-ray diffraction spectrum using $\text{CuK}\alpha$ -ray, and has a specific surface area of from 2 to $10 \text{ m}^2/\text{g}$.

14. The production process according to any one of

5 Claims 10 to 13, wherein the cobalt hydroxide or the tricobalt tetraoxide has a press density of from 1.2 to 2.5 g/cm^3 .

15. The production process according to any one of Claims 10 to 14, wherein the lithium-cobalt composite

10 oxide has a half value width of the diffraction peak on (110) plane of from 0.07 to 0.14° , a specific surface area of from 0.3 to $0.7 \text{ m}^2/\text{g}$, a heat generation starting temperature of at least 160°C , and a press density of from 3.1 to 3.4 g/cm^3 .

15 16. A positive electrode for a lithium secondary battery, which contains the positive electrode active material as defined in any one of Claims 1 to 5.

17. A positive electrode for a lithium secondary battery, which contains a positive electrode active material
20 obtained by the production process as defined in any one of Claims 6 to 15.

18. A lithium secondary battery employing the positive electrode active material as defined in Claim 16 or 17.